3. Apatite Fission-Track Analysis

3.1 Introduction

Apatite fission-track (AFT) analysis has been widely used during the past 30+ years to constrain the low-temperature thermal histories of many igneous, metamorphic and sedimentary rocks in a wide range of geological settings. Applicable geological settings include orogenic belts, rifted margins, faults, sedimentary basins, cratons, and mineral deposits. The types of geologic problems that can be addressed include the timing and rates of tectonic events, sedimentary basin evolution, the timing of hydrocarbon generation and ore mineralization, the absolute age of volcanic deposits, the effects of major climatic changes on the near-surface geothermal gradient, and long-term landscape evolution. Early work by Naeser (1967) and Wagner (1968, 1969) first established the basic procedures that enabled the fission-track dating method to be applied routinely to these geologic problems. Fleischer et al. (1975) summarized the early studies of the broader discipline of nuclear-track detection in different solid-state materials. More recent comprehensive reviews of fission-track applications have been provided by Naeser and McCulloh (1989), Wagner and Van den haute (1992), Gallagher et al. (1998), Van den haute and De Corte (1998), Dumitru (2000), and Gleadow et al. (2002). Etched, natural fission tracks in several apatite grains are shown in Figure 1. Successful AFT analysis is limited by the following: 1) the availability of apatite from which useful AFT data can be obtained, often due to a lack of apatite of sufficient grain size and quality within available rock types, or, alternatively, the lack of available rock samples due to minimal outcrop exposure or other reasons, 2) economic considerations in terms of the time and money required to obtain sufficient AFT data, 3) the inherent limitations of AFT data to resolve geological thermal history information, often related to limited numbers of accumulated spontaneous fission tracks due to low uranium concentration within the available apatite and/or young apatite grain fission-track ages, 4) the limitations of the model(s) upon which AFT interpretations are based, and 5) self-imposed limitations by various laboratories, often in the form of the measurement and interpretative schemes employed.
The recent resurgence of (U-Th)/He dating, in particular for the minerals apatite, zircon, and titanite (e.g., Reiners 2003 and references therein), complements and challenges AFT methods. The complementary aspect resides in the low-temperature nature of the various (U-Th)/He systems. The challenge resides in the periodically encountered, apparent incompatibility of the data derived from the various techniques (e.g., apatite (U-Th)/He ages > AFT ages). Incompatible (U-Th)/He and fission-track data demonstrate that the systematics of one or both methods are incompletely understood. Therefore, considerable study and debate in the AFT community now centers on the type of data to measure, which methods are used to collect those data, and which models are developed and employed to interpret those data in terms of geological thermal histories.
3.2 Apatite as a Fission-Track Analysis Material

3.2.1 General

Several fundamental characteristics of apatite make it an excellent mineral for fission-track analysis and provide the basis for the AFT methods presented here. These are: 1) its nearly ubiquitous natural occurrence in many common crustal rock types, 2) its physical properties, 3) its major and minor element chemistries, 4) its trace element chemistry, in particular the presence of uranium and thorium in most natural crystals, 5) its ability to retain fission tracks in the geological environment, and 6) the ability of an experimentalist to mimic important features of the behavior of spontaneous fission tracks in the geological environment using laboratory analogues.
3.2.2. Natural Occurrence

Apatite is a nearly ubiquitous accessory mineral, found in many types of igneous, metamorphic, and sedimentary rocks. As used throughout this discussion, an apatite grain or simply a grain may be a whole apatite crystal, a fragment of an apatite crystal, or an assemblage of multiple whole crystals and/or crystal fragments that may or may not have experienced a sedimentary transport history. Apatite grains composed of whole crystals or crystal fragments occur as: 1) anhedral to euhedral accessory grains in igneous rocks, 2) detrital grains in clastic sedimentary rocks, 3) primary and secondary grains in shales, limestones, and coals, and 4) primary mineral grains or porphyroblasts in metamorphic rocks including marbles. Apatite grains composed of aggregate crystals occur as: 1) biologically-derived apatite or phosphatic material including teeth, bones, and conodonts, 2) chemical precipitates or nodules in phosphate-rich sedimentary rocks, and 3) diagenetic or hydrothermal grains in clastic and non-clastic sedimentary rocks. Apatite usually occurs as small sand-sized grains, typically < 300 µm (< medium sand or < phi scale value of +1 to +2) across the short diameter, in amounts less than 1% of the total rock volume. AFT analysis methods generally require apatite grains to have a minimum diameter of approximately 50 µm (≥ very fine sand or ≥ phi scale value of approximately +4), although smaller grains can be used.
3.2.3 Physical Properties

Most naturally occurring apatite is a member of the $6/m$-hexagonal dipyramidal crystal class (Deer et al. 1969); the known exception being the uncommon, near-end-member chlorapatite which is monoclinic (e.g., Hughes et al. 1989). The typical crystal habit of apatite derived from igneous rocks is prismatic. In a sedimentary transport system, such primary accessory apatite grains usually experience some degree of rounding of the crystal edges. In sandstones, any degree of rounding may be evidenced from undetectable to complete, but it is somewhat uncommon to find apatite grains that are completely rounded into a nearly spherical shape. Apatite possesses a weak parting perpendicular to the crystallographic $c$-axis, but possesses no strong cleavage. Furthermore, apatite is soluble in acidic aqueous solution and except for the rounding effects mentioned above and possible interaction with natural acidic solutions (e.g., humic acid in tropical weathering regimes) apatite commonly survives surface and near-surface geological processes. Apatite has a specific gravity of between 3.15-3.20 g/cm$^3$, making it a heavy mineral relative to quartz, feldspar, calcite, and other rock-forming minerals that tend to have specific gravities less than 2.90 g/cm$^3$. In a standard 30 µm thick thin section, apatite appears as first order gray or white (perpendicular to the $c$-axis) and can be distinguished from quartz based on its greater relief due to its higher refractive index (~1.63) compared to that of quartz (1.54). In hand specimen, apatite is typically difficult to impossible to identify, even with a hand lens, due to its generally low abundance level.
3.2.4 Major and Minor Element Chemistries

Apatite is the most common phosphorous-bearing mineral in the Earth’s crust. The chemical formula for apatite is generally written as Ca$_5$(PO$_4$)$_3$[F,Cl,OH] (Deer et al. 1969). The three anions F$^-$, Cl$^-$, and OH$^-$ are believed to substitute for one another between the various end-member compositions fluorapatite, chlorapatite, and hydroxyapatite, respectively. Appreciable CO$_2$ is known to occur in some natural apatites. Additional substitutions include Mn, Sr, Fe, Na, and rare-earth elements (particularly Ce) in the Ca-site and Si, S, and C in the P-site (Deer et al. 1969; Young et al. 1969; Roeder et al. 1987). Near-end-member calciamfluorapatite is the dominant apatite variety in most crustal rocks. However, it is quite common for relatively non-fluorapatite compositions to be found in crustal rocks, especially in sandstones that contain apatite grains from multiple provenance sources.
3.2.5 Uranium and Thorium as Trace Elements

Uranium and thorium are important trace elements in apatite, the former being important for AFT analysis and both being important for apatite (U-Th)/He analysis. The concentration of natural uranium in apatite typically ranges from 1-200 ppm. Under oxidizing conditions, uranium forms the uranyl ion (UO$_2^{+2}$), which in turn forms compounds that are soluble in aqueous solution. Because uranium in apatite is protected from the environment by its host crystal lattice, it is not lost from the host grain when the apatite is immersed in water, but it can be lost by a process of dissolution and re-precipitation. The combination of the high charge and relatively small ionic radius of the uranium cation ($U^{+4} = 1.05$ Å) makes uranium incompatible with other cations in the common silicate minerals. Consequently, uranium tends to be concentrated in accessory mineral phases such as apatite, zircon, titanite, and monazite. Natural uranium is composed of the three isotopes $^{234}$U, $^{235}$U, and $^{238}$U. The ratio $^{238}$U/$^{235}$U of natural uranium is adopted by convention (Steiger and Jäger 1977) to be exactly 137.88. Natural uranium is distinguished from enriched uranium (enriched in $^{235}$U; depleted of $^{238}$U) or depleted uranium (depleted of $^{235}$U; enriched in $^{238}$U), enriched and depleted uranium being dominantly the result of human processing of natural uranium for fuel or weapons. A summary of the natural uranium and thorium isotopes, including their relative abundances and nuclear properties relevant to AFT analysis, is given in Table 1. Nuclear fission of the isotope $^{238}$U produces the spontaneous fission tracks in apatite. The isotope $^{235}$U, on the other hand, is important because it is often used to measure the amount of $^{238}$U present in an apatite grain. Additionally, $^{235}$U is used extensively in laboratory-based, experimental studies of fission tracks in apatite because it can be easily induced to form fission tracks under carefully controlled conditions (see below). $^{238}$U experiences spontaneous nuclear decay by two processes: α-decay (emission of a $^4$He nucleus), the dominant process, and spontaneous fission (splitting of the nucleus into two fragment nuclei, rarely three, plus 2 or 3 high-energy neutrons; Friedlander et al. 1981). The $^{238}$U α-decay process is composed of a complicated series of individual α-particle emitting nuclear reactions that ultimately lead to a stable $^{206}$Pb nucleus. For every approximately two million $^{238}$U nuclei that undergo α-decay, a single $^{238}$U nucleus will experience spontaneous nuclear fission. For purposes of AFT analysis, the large thermal-neutron
capture cross-section of 235U relative to those for 234U, 238U, and 232-Th is of great importance (see Table 1). Thermal neutrons are neutrons traveling at speeds similar to the speeds traveled by gas molecules at room temperature (approximately 2200 m s\(^{-1}\)). Thermal neutrons are available from the core of a nuclear fission reactor that is surrounded by a medium (such as light or heavy water with a high spatial density of hydrogen or deuterium nuclei, respectively) that moderates the velocities of the neutrons emanating from the core. The thermal-neutron capture cross-section is the effective target area that a nucleus presents to a thermal neutron. By exposing a natural mixture of uranium and thorium to a flux of thermal neutrons, it is possible to induce fission of 235U with negligible induced fission of the other uranium and thorium isotopes. The process of inducing fission of 235U in this manner permits 235U to be used as a means of determining the amount of 238U present in an apatite grain because the ratio of 238U to 235U is a fixed number in most natural materials (Steiger and Jäger 1977).

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Abundance</th>
<th>Half-Life</th>
<th>Decay Constant</th>
<th>Thermal-Neutron Capture Cross-Section ((\sigma))</th>
</tr>
</thead>
<tbody>
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<td>235U</td>
<td>100.0000</td>
<td>1.41 × 1010 ((\alpha))</td>
<td>4.916 × 10^-11 ((\alpha))</td>
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<td>0.0037</td>
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<td>9.8485 × 10^-10 ((\alpha))</td>
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<tr>
<td>232U</td>
<td>99.2743</td>
<td>4.468 × 109 ((\alpha))</td>
<td>1.55125 × 10^-10 ((\alpha))</td>
<td>2.7</td>
</tr>
<tr>
<td></td>
<td>-1.3 × 1016 (s.f.)</td>
<td>-7.5 × 10^-17 (s.f.)</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

NOTES: (\(\alpha\)) indicates alpha-decay series, (s.f.) indicates spontaneous fission-decay.

Table 3.1
3.3 Formation and Registration of Nuclear Fission Tracks

3.3.1 Spontaneous and Induced Nuclear Fission Decay

Nuclear fission is a process during which a heavy, unstable nucleus splits into a pair of fragments of similar size. This reaction takes place both spontaneously in nature and artificially during bombardment by neutrons and other high-energy particles or γ-rays. Each reaction is accompanied by the release of a few neutrons and ~210 MeV of energy, of which the majority (~170 MeV on average) is the kinetic energy of the fission fragments. As a result of this energetic disintegration, the fission fragments with massive positive charges are propelled from the reaction site in opposite directions. If the nucleus is located within a dielectric solid, the reaction will create a damage trail, called a fission track, along the trajectories of the two fragments.

Spontaneous fission occurs in very heavy nuclides that belong to the actinide series of elements. Of those nuclides, $^{232}$Th and three U isotopes ($^{234}$U, $^{235}$U and $^{238}$U) are the typical candidates that produce a significant number of spontaneous fission tracks in solids. However, in regards to the relative abundances and spontaneous fission half lives, $^{238}$U is the only source of spontaneous tracks in terrestrial materials including natural apatite and zircon, except for those anomalously enriched in Th (Wagner and Van den haute 1992). Note that $^{238}$U also decays through a chain comprised of eight α ($^4$He) and six β− emissions. This decay, which occurs $2 \times 10^6$ times more frequently than the spontaneous fission events, needs to be taken into account in calculating an FT age. This will be discussed in more detail later.
3.3.2. Track Formation Process in Solids

When a heavy ionized particle travels at a high velocity through a solid, it interacts with the host lattice, gradually loses its kinetic energy and slows down until it eventually stops. During this process, a couple of interactions are predominantly responsible for the deceleration of the particle: (1) electric interaction, more effective at high velocities, in which the particle surrenders its energy by stripping electrons from target atoms, by having its own electrons stripped away, and by raising the excitation level of the lattice electrons; and (2) nuclear interaction, more important as the particle slows down, in which the particle loses its energy by elastic collisions with lattice atoms. In addition, the energy is also partitioned to radiation (e.g., Chadderton 2003). Although there is good agreement on the deceleration process of the charged particle and induced interactions, the debate has raged as to which mechanism causes the resultant motion of the lattice atoms that forms and registers the damage trails (e.g. Fleischer et al. 1975; Durrani and Bull 1987; Wagner and Van den haute 1992; Gleadow et al. 2002; Chadderton 2003). One of the widely accepted theories is the “ion explosion spike” model (Fleischer et al. 1965a, 1975) that treats electrostatic displacements as the primary process. In this model, track formation occurs during three stages (Fig. 1): (1) the rapidly moving positively-charged particle strips lattice electrons along its trajectory, leaving an array of positively-ionized lattice atoms; (2) the resulting clusters of positive ions are displaced from their original lattice sites as a result of Coulomb repulsion, creating interstitials and vacancies; and (3) the stressed region relaxes elastically, straining the surrounding undamaged lattice. The creation of the long-ranged strains in the third stage makes possible the direct observation of unetched (or latent) tracks by transmission electron microscopy (TEM). This model explains the primary observation that particle tracks are observed only in dielectric solids having electric resistivity >2000 Ωm (Fleischer et al. 1975). An alternative is the “thermal spike” model (Seitz 1949; Bonfiglioli et al. 1961; Chadderton and Montagu-Pollock 1963), in which the passage of an energetic particle is assumed to produce instantaneous, intense heating of the lattice along the trajectory. The track core is rapidly heated to a high temperature and subsequently quenched by thermal conduction into the surrounding lattice. As a result, lattice defects are created by the thermal activation and the track core is left disordered. Chadderton (2003)
argued that both ion explosion and thermal spikes could be present to different degrees for track formation and registration in a variety of solids.
3.3.3 Structure of the Latent Track

Knowledge on the structure of fission tracks in their latent state is critical not only to unravel their formation process but also to serve as a physical basis for their thermal annealing behaviors, which have conventionally been observed and quantified after chemical etching. Since the pioneering work by Silk and Barnes (1959), the atomic-scale characterization of latent tracks has been conducted using TEM and other analytical techniques on apatite (Paul and Fitzgerald 1992; Paul 1993), zircon (Yada et al. 1981, 1987; Bursill and Braunshausen 1990), muscovite mica (Thibaudau et al. 1991; Vetter et al. 1998) and other dielectric materials (see overviews by Fleischer et al. 1975; Wagner and Van den haute 1992; Neumann 2000; Chadderton 2003) (Fig. 2).

In general, latent tracks represent a cylindrical shape of amorphous material in a crystalline matrix, with a sharp amorphous-crystalline transition. The elastic strain field around the disordered core extends a short distance into the matrix, likely with $1/R^2$ dependence (Bursill and Braunshausen 1990; R, radial distance), with no evidence for structural defects in its vicinity. The cross section of the track has a nearly circular shape with widths of 6-10 nm in apatite (Paul 1993), ~8 nm in zircon (Bursill and Braunshausen 1990) and ~4-10 nm in muscovite mica, depending on the ion energy loss (Vetter et al. 1998). The track is a linear, continuous feature and has approximately
Figure 3.2: Formation and registration process of charged particle tracks in a dielectric solid by the ion explosion spike model (Fleischer et al. 1965a), which involves three stages: (A) the rapidly moving positively-charged particle strips lattice electrons along its trajectory, leaving an array of positively-ionized lattice atoms; (B) the resulting clusters of positive ions are displaced from their original lattice sites as a result of Coulomb repulsion, creating interstitials and vacancies; and (C) the stressed region relaxes elastically, straining the surrounding undamaged lattice.
When combined with the apparent fission track age, length distributions can be used to reconstruct the variation of temperature through time. Observations of track lengths from a wide variety of surface rocks (Gleadow et al. 1986) show that distinctive patterns of track length characterize particular geological environments. Figure 10 illustrates the major length distribution categories identified by Gleadow et al. (1986) in rocks for which the thermal history is known, or can be inferred with reasonable accuracy. The undisturbed volcanic type is, as its name implies, characteristic of volcanic rocks that have remained undisturbed and at relatively low surface temperatures since their formation. A similar pattern will result in any rock which has cooled rapidly and not been re-heated thereafter. This type of distribution is similar to that shown by fresh induced tracks, although the mean length is slightly lower, indicating that some shortening occurs in spontaneous tracks even at ambient surface temperatures. Apatite grains that have spent a significant period of time within the fission track annealing zone will show various patterns of broader length distribution, such as the undisturbed basement type, representing monotonic cooling from temperatures above about 120°C. More complex, multi-stage thermal histories will produce the even broader ‘mixed’ distributions. When the peaks in such a distribution are clearly resolved, as in the bimodal case, the distribution is indicative of a two-stage history with an older generation of tracks shortened during a later thermal event, and a new generation of long tracks produced subsequently. Such a bimodal distribution is particularly useful, giving information on the timing as well as the severity of the thermal event.
3.4 Track Stability

The phenomenon of fission track fading, or annealing, was first recognized by Silk and Barnes (1959) who showed that materials that had undergone heating contained shorter tracks than untreated specimens. The fading mechanism was initially interpreted by Fleischer et al. (1964) as a pinching out of segments along the track which prevented further access of the track etchant. Naeser (1979a) suggested that the diffusion rate within a heavily annealed track gradually approached that of the undamaged solid, so that it became increasingly difficult to etch out the track. It remains unclear precisely how the structure of the track, at the atomic level, may be altered in response to changes in environmental conditions, such as pressure, ionization, increased temperature, and duration of exposure (Fleisher et al. 1965b), but it is now generally accepted that temperature and time are the primary controls on track stability. Experimental studies by Green et al. (1986) confirmed that track fading was probably a two-part process, in which tracks at first begin to shrink from each end, while continuing to be etchable for the remainder of their length. Eventually track fading enters another phase, one of segmentation (Green et al. 1986), where the latent track cannot be fully etched and is apparently broken by small unetchable gaps (see also Hejl 1995). Transmission Electron Microscope observations of highly annealed tracks have provided additional evidence for unetchable gaps in highly annealed tracks (Paul and Fitzgerald 1992).

![Figure 3.4](image)

Figure 3.4 Representative track length distributions for spontaneous tracks in the
various apatite length groups recognized by Gleadow et al. (1986). The top row represents measurements on horizontal confined tracks for which the differences between the different types are more distinctive than for the corresponding projected length distributions (bottom row). 100 confined tracks and 500 projected track lengths were measured in each case. After Gleadow et al. (1986).

A fission track has its inception when two fission fragments pass violently through the crystal lattice at high velocities with an initial energy of around 1 MeV/nucleon (Fleischer et al. 1975). The metastable damage zone formed (Fig. 2) immediately begins to heal (Green 1980, Donelick et al. 1990) at a rate largely determined by the temperature of the sample and, to a lesser extent, the duration of the elevated temperature (Laslett et al. 1987). The tracks will continue to shorten until they cool to lower temperatures. The final length of each track therefore represents the integrated result of its passage through time-temperature so that each, in effect, behaves as maximum-recording thermometer. Progressive shortening of the confined track length is accompanied by a reduction in the measured track density. This is due to the reduced probability of shortened tracks intersecting the polished surface of a grain and thus being exposed to the etchant. As the observed or apparent age of samples is determined on the basis of track density (Naeser and Faul 1969, Wagner and Reimer 1972, Nagpaul et al. 1974) considerable research Fission Track Dating of Phosphates, Thermochronology of Apatite 595 aimed at determining the reduction of track density during annealing has been reported (Wagner and Storzer 1972, Bertel and Mark 1983, Laslett et al. 1984, Green 1988). The term “apparent” is emphasized since the observed age may be modified by the amount of track annealing that has occurred.

Variations in the observed fission track age with track fading initially seemed to be something of an impediment to mineral dating by this method. However, the systematic and progressive nature of the track length reduction presented an unparalleled opportunity to extract thermal history information in addition to the chronology (Wagner and Storzer 1972). Track length distributions observed in geological samples, were seen to be the net result of both track production and track fading processes over a span of geological time. Indeed, it was clear that there was a
wealth of information available if the natural track distributions could be understood.

3.4.1 Annealing Over Geological Time-Scales

Typical crustal geothermal gradients are around 20-30°C/km so that the temperature at 4-5 km depth is the range 100-120°C, allowing for surface temperatures of around 10-20°C (Pollack et al. 1993). The analysis of samples from deep bore holes (e.g., Naeser and Forbes 1976, Naeser 1981, Gleadow and Duddy 1981, Hammerschmidt et al. 1984), has provided direct evidence of natural thermal annealing of fission tracks in apatite over geological time-scales. Data from hydrocarbon exploration wells drilled within the Otway basin in southeastern Australia (Gleadow et al. 1983, Green et al. 1989a) clearly demonstrate a systematic reduction in the mean confined track length and apparent fission track age with increasing temperature (Fig. 11). At temperatures greater than about 120°C (depths >~3km) no fission tracks are preserved within apatite and so the apparent fission track age and mean length are effectively zero. Both the apatite fission track age and mean track length decrease systematically with depth from the initial provenance age of ~125 Myr at the surface to zero at a depth of ~3.5 km forming a characteristic concave-up profile of apparent apatite age (Fig. 11).
Figure 3.5: Composite apatite fission track crustal profiles of mean fission track length (•) and apparent apatite fission track age (○) plotted against depth for samples from several wells from the central Otway Basin in southeastern Australia. These clearly illustrate the progressive decrease in mean track length and apparent apatite fission track age with depth, and the characteristic concave up form of both profiles. After Gleadow and Duddy (1981b) and Green et al. (1989a).

Four representative track length distributions for different depths in these wells are shown in Figure 12. The observed increase in the standard deviation of the length distribution with decreasing mean track length (higher T) is a consequence of the anisotropy of track shortening (tracks perpendicular to the c-crystallographic axis anneal faster than tracks parallel to the c-axis) as well as the variation in apatite composition between grains (e.g., Green et al. 1985, Green 1988). The thermal history
for Otway basin samples can be reconstructed from the relatively simple burial history and indicates that this pattern was produced by heating times in the range of 10-100 Myr. Also shown in Figure 12 are a series of histograms of single grain ages with smoothed probability distributions and radial plots (Galbraith 1990) from the same four samples (Dumitru et al. 1991, Gallagher et al. 1998).

Other examples of natural thermal annealing can be seen in the vicinity of shallow level igneous intrusions. Calk and Naeser (1972), for example, demonstrated a systematic reduction in the apparent apatite fission track age of an 80 Ma old granitic pluton with increasing proximity to the contact with a small (~100 m) basaltic intrusion emplaced ~10 Ma ago. This pattern of age reduction within the granite was influenced by the thermal effect of the basalt intrusion and is consistent with the pattern of annealing observed in laboratory annealing experiments and deep drill holes. In order to improve our understanding of the relationships between time, temperature and the observed track parameters in the natural environment, numerous experiments have been conducted over the last twenty-five years. Perhaps the most useful outcome of these annealing experiments has been the development of robust mathematical modeling. Ever-more sophisticated models continue to be developed, from which geological interpretations of greater precision can be extracted. We will now examine some of the laboratory annealing experiments, as well as some of the current ideas on the processes involved, highlighting some the strengths and the weaknesses of the approach, and describing several problems that have been overcome to make the fission track technique a versatile and powerful tool in tectonic and landscape analysis. This review of some of the key developments in apatite annealing as it pertains to fission track thermochronology includes a selective and by no means exhaustive bibliography.
3.4.3 Fission Track Annealing

Fission tracks shorten irreversibly, or anneal, when the host mineral is at an elevated temperature, as a consequence of a thermally activated diffusion process that is poorly understood. The temperature range over which annealing occurs varies for different minerals and also depends on the heating rate (see Appendix B of Wagner & Van den Haute 1992). The most commonly analyzed minerals are apatite, zircon, and sphene, with the emphasis on the first two. We only consider these two here and focus specifically on apatite. The concept of a closure temperature (e.g. Dodson 1973) for the fission track system, i.e. the temperature of the system at the time given by its apparent age, is generally an oversimplification. For example, 110 ± 10°C is commonly adopted for the nominal closure temperature of apatite, yet geological examples indicate that significant annealing occurs over a temperature range of at least 60°C (e.g. Gleadow & Duddy 1981), and this range is generally referred to as the partial annealing zone, or PAZ (e.g. Wagner 1979b). The 60°C range for apatite is a minimum range, as it is well known that the mean of a distribution of induced tracks in apatite is 1-1.5 µm longer than spontaneous tracks measured from samples that are considered to have been at near-surface temperatures since their formation (e.g. Green 1988). This suggests that some annealing occurs at near-surface temperatures over geological time scales. Donelick et al (1990) demonstrated that some annealing of induced tracks in apatite occurs at room temperature over time scales of minutes to a few weeks, although the length of tracks seems to stabilize after this. Overall, the rate of annealing depends on temperature, time, and the track length through a strongly nonlinear relationship (e.g. Laslett et al 1987, Carlson 1990, Crowley et al 1991, Laslett & Galbraith 1996), and it is likely that the dominant process changes as annealing progresses.
3.4.3 Annealing Characteristics of Apatite

A landmark for the application of apatite fission track data to geological problems was a comprehensive suite of well-controlled laboratory experiments, which were conducted to constrain the annealing characteristics of track lengths in apatite, in particular Durango apatite from Mexico (Green et al 1986, 1989a,b, Laslett et al 1987, Duddy et al 1988). The experiments were performed under both isothermal and variable temperature conditions, with heating durations between 20 min and 500 days and temperatures between 95˚ and 400˚C. The basic outcome of this work was a quantitative, albeit empirical, predictive model of fission track annealing, expressed in terms of the reduction in track length for a given temperature over a particular time interval. The basic formulation is an Arrhenius-type model (i.e. dependent on inverse absolute temperature and log of time) and is given by Laslett et al (1987). Extrapolation of Laslett et al's (1987) Durango apatite model to geological time scales (106 to 108 years) predicts a partial annealing temperature range from ~60˚ to ~110˚C, with an uncertainty of about 10˚C. Apatite composition may be an important control on annealing rate, particularly the relative proportions of Cl, F, and OH. It has been known for some time that Cl-apatite seems to be more resistant to annealing than F-apatite (Gleadow & Duddy 1981, Green et al 1986, Donelick 1991, O’Sullivan & Parrish 1995). Crowley et al (1991) have presented results of laboratory annealing experiments on F-apatite. Extrapolation of their preferred annealing model to geological time scales predicts F-apatite to be more resistant than the more Cl-rich Durango apatite (F/Cl ratio of ~0.1; Young et al 1969), in contrast to the geological observations. More recently, Laslett & Galbraith (1996) used a slightly different model parameterization than Crowley et al's (1991), and this new model is consistent with the general observation that F-apatite is expected to anneal more easily than the Durango apatite. Relatively little is known about the role of other species such as OH and Mn (Ravenhurst et al 1992, Bergman & Corrigan 1996). Burtner et al (1994) have presented data relating the degree of annealing to Cl composition and the etching characteristics of apatite (quantified in terms of the size and geometry of etch pits). Recently, RA Donelick & RA Ketcham (personal communication) have developed an annealing model incorporating these factors, although this has not been published at the time of writing. Barbarand et al (1997),
using cathodoluminescence spectroscopy, have recently identified an apparent control in terms of lattice structure rather than F/Cl composition. They argue that there are controls in addition to the compositional effects, such as crystallographic imperfections, and these are reflected in the etch pit geometries. It is still a matter of debate as to which annealing algorithm is most appropriate, and it is likely that no single model will be universally applicable. Currently, it is difficult to assess the importance of apatite chemistry and crystal structure quantitatively, and sufficient details of these factors, which may allow them to be incorporated into a single generalized annealing model, have not yet been published.

<table>
<thead>
<tr>
<th></th>
<th>1 hr.</th>
<th>1 m.y.</th>
<th>100 m.y.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Apatite</td>
<td>345</td>
<td>125</td>
<td>100</td>
</tr>
<tr>
<td>Biotite</td>
<td>415</td>
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<td>350</td>
</tr>
<tr>
<td>Tourmaline</td>
<td>585</td>
<td>295</td>
<td>960</td>
</tr>
<tr>
<td>Zircon</td>
<td>720</td>
<td>380</td>
<td>340</td>
</tr>
</tbody>
</table>

Table 3.6
3.4.4 Partial Annealing Zone (PAZ)

Annealing of fission tracks on minerals does not occur at a single temperature, yet if represents a wide temperature range. A variation of the annealing rate which is likely to occur due to chemical composition has to be considered (Wagner and Vanden Haute, 1992). The range of temperatures with respect to trace stability is divided by three zones (Wagner, 1972). Zone 1, is called total annealing which occur at high temperatures. In this zone no tract forms, hence the fission track age in zero. At medium temperature, the tracks are more stable with the degree of stability between 0 until 1. In this stage, which is known as the partial annealing zone (Zone II), the tracks are partially annealed. The partial annealing zone (PAZ) is of great importance whereas the fission track age starts accumulating at this moment. At low temperatures, the tracks are stable (1 degree of stability). Samples residing in this stage consist of unannealed tracks which formed and stay within this zone (Zone III) and tracks which have been partially annealed (Zone III). All of the zones are illustrated below:

In reality, the PAZ concept cannot be straightforward applied. The lowermost temperature of the PAZ is easy to determine whereas the tracks are completely extinct. However, the PAZ top is more difficult to be defined. In apatite for instance, tracks cannot reach a complete stability at surface temperature. Therefore, the PAZ is defined as the gradient of stability reaches its maximum, namely 0.90 - 0.95. On the other hand the rate of geological process also gives the annealing temperature
differently. A fast process, such as volcanism which involved a rapid change of temperature, will need a higher temperature for tracks to be annealed (Wagner and Van den Haute, 1992). The temperature required to anneal is lower for geological process such as cooling of crystalline basement, uplift and denudation (Gleadow and Lovering, 1978b). Besides of wide annealed temperature range (PAZ), a single temperature value is often applied for age interpretation. It is called the closure temperature. The experiment conducted on Durango apatite by Laslett et al. (1987), which was performed under a given temperature range and the time interval resulted that the PAZ of apatite ranges from ~60°C to ~110°C, with an uncertainty of about 10°C.
3.4.5 Closing Temperature and Thermal History

The closing temperature $T_c$ can be given a simple and precise definition namely, the temperature of the system at the time given by its apparent age. According to Wagner (1972) the fission track age can be taken to represent the time when the rock cools down to a temperature at which 50% of the tracks are stable. Laboratory annealing data can be extrapolated to the times of the order of $10^7$ - $10^8$ years to find the temperature at which 50% of the tracks are stable. This gives the losing temperature. In volcanic rocks cooling takes place very rapidly and different co-existing minerals in them will give the same fission tracks ages. But in rocks like granites which generally cool slowly the different co-existing minerals having different closing temperatures will act like temperature recorders.

The cooling rate will be given by

$$\text{Cooling rate} = \frac{T_1 - T_2}{A_1 - A_2}$$

Where $T_1$, $T_2$ are closing temperatures and $A_1$, $A_2$ are the ages of two minerals. The temperature of the earth increases by 30°C per km of depth (average geothermal gradient). If a granite be uplifted by 5 km. it will reach at height cooler by 150°C. Thus a cooling by 150°C way mean an uplift of 5 km. Thus we find that Assuming that the cooling has taken place due to uplift alone.

$$\text{Uplift} = \frac{\text{Cooling rate}}{\text{Geothermal gradient}}$$

$$\text{Uplift rate} = \frac{\text{Cooling rate}}{\text{Geothermal gradient}}$$
3.4.7. Application of Apatite Annealing Models to Geological Time Scales

One of the problems inherent in current annealing models for apatite (beyond the chemistry and crystallographic issues) is the extrapolation of laboratory measurements to geological time scales. Some geological studies are used to justify the models a posteriori, but these arguments are somewhat unsatisfactory, as it is necessary to assume the geological thermal history. Consequently, some debate still surrounds the details of which model is the most acceptable (Vrojlik et al. 1992, Carlson 1993a,b, Corrigan 1993, Crowley 1993a, Green et al. 1993, Burtner et al. 1994, Bergman & Corrigan 1996, Green 1996, Barbarand et al. 1997). Accepting these limitations, predictive annealing models for apatite do allow quantitative reconstruction of thermal histories in the upper several kilometers of the Earth's crust [i.e. up to temperatures of 120°-130°C on time scales of 1-100 million years (My)]. These reconstructions can be generated by forward modeling (e.g. Duddy et al. 1988, Green et al. 1989ab, Willet 1992, Crowley 1993b) or, preferably, by some optimization procedure using the observed data as constraints (e.g. Corrigan 1991, Lutz & Omar 1991, Gallagher 1995, Willet 1997). A uniform diameter along most of its length. For its entire range, however, the track is a cylinder over a certain length and has a tapering-down in diameter near its end (Dunlap et al. 1997; Chadderton 2003), which may be the case of both terminals of a fission track (Carlson 1990).

The axial variation in damage density is deduced from a theoretical consideration on the rate of kinetic energy loss along a trajectory of nuclear fission fragments (Green et al. 1988; Wagner and Van den haute 1992). Since the kinetic energies of a pair of fission fragments are both below the Bragg peak in the energy loss curve, both fragments are most intensely ionizing, and therefore create most intensive damages, in the early part of their passage through the crystal lattice, i.e., around the site of the nuclear fission. As they slow down and lose energy, the damage intensity falls off away from this point, with the result that the damage density along the track peaks around the center and falls to zero at each end (Green et al. 1988).
Figure 3.7: Atomic-scale images of latent (unetched) tracks: (A) an induced track in Durango fluorapatite observed subparallel to its length by transmission electron microscopy (Paul and Fitzgerald 1992) and (B) intersections of Pb ion tracks in muscovite mica imaged by 400 kV high-resolution transmission electron microscopy (Vetter et al. 1998).
3.5 Chemical Etching and Optical Microscope Observation

Latent fission tracks can be viewed only by TEM and other high-resolution microscopic techniques because of their limited widths of only several nm. They, however, are conveniently scanned only when samples have very high track densities (i.e., > $10^{10}$ cm$^{-2}$), which is generally not the case of terrestrial minerals such as apatite and zircon. Another potential problem with viewing latent fission tracks is that the entire range of each track cannot be observed at such a high magnification, despite the necessity to analyze track lengths for the modern FT thermochronometry method (Paul 1993). In this regard, a technique is needed to enlarge fission tracks in order to be visible at a lower magnification under an optical microscope of the visualization techniques available, by far the most general and widely used has been chemical etching, which utilizes the preferential chemical attack along the entire length of the fission tracks.

3.5.1 Basic process of Track Etching

Fission tracks can be enlarged by chemical etching because the disordered region of the track core is more rapidly dissolved than the surrounding undamaged bulk material, due to its lowered binding energy. Chemical etching is conducted by immersing a dielectric material into a particular reagent under strictly controlled temperature and time conditions. Thus, only tracks that intersect the material’s surface are etched and enlarged for observation. The reagent and etching conditions are selected on an empirical basis and have been established for a variety of minerals and other dielectric solids (Fleischer et al. 1975; Durrani and Bull 1987; Wagner and Van den haute 1992). The geometry of an etched track is controlled by the simultaneous action of two etching processes: chemical dissolution along the track at a rate $V_T$ and general attack on the etched surface and on the internal surface of the etched track at a lesser rate $V_G$ (Fleischer and Price 1963a,b) (Fig. 3). Under given etching conditions, $V_T$ in general increases with ionization rate and thus varies along a track, whereas $V_G$ is generally constant for a given material but can depend on crystallographic orientations. In natural glasses, which are typically amorphous, $V_T$ is only a few times higher than $V_G$ and hence the etched tracks appear as rounded cones. In crystals, however, $V_T$ is generally much higher than $V_G$ by more than a factor of 10 and this results in a needle-like track shape (Fig. 4). In
the course of such etching, dissolution and removal of the latent track core is completed before the track is sufficiently enlarged and becomes visible under the optical microscope. Therefore, the visible enlargement process is predominantly governed by the progressive widening of the track channel to \( \sim 1 \, \mu m \) at a rate \( V_G \).

Figure. 3.8 : (A) The geometry of an etched track is controlled by two etching processes, i.e., chemical dissolution along the track at a rate \( V_T \) and general attack on the etched surface and on the internal surface of the etched track at a lesser rate \( V_G \) (Fleischer and Price 1963a,b): (B) Tracks inclined at relatively low angles to a surface (i.e., less than the critical angle, \( \theta_c \)) are not etched to be observable under optical microscope: (C) New tracks that began and ended beneath the original surface are revealed by progressive removal of the surface itself. Here \( V_G/V_T \) is given as 1/3 throughout.
Figure 3.9: Photographs of etched fission tracks viewed under optical microscope. (A) Spontaneous tracks revealed on a polished internal surface of ~27.8 Ma Fish Canyon Tuff zircon. The crystallographic c-axis lies approximately vertical. (B) Induced tracks implanted on a muscovite detector (Brazilian Ruby clear) that were derived from the region of the photograph (A). (Photos by TT) (C) Spontaneous tracks on a polished internal surface of ~33 Ma apatite crystal. The c-axis lies approximately horizontal. (Photo by POS) Scale bars are 10 µm.
3.5.2 Etching Efficiency and Prolonged-Etching Factor

Because the surface of the mineral grain or glass is attacked and progressively removed during etching, tracks inclined at relatively low angles to the original surface are etched away. Such low-angle tracks are therefore not observable under the optical microscope (Fig. 3), which results in the number of latent tracks intersecting a given surface no longer being equal to that of etched ones on the surface. The minimum angle to the surface above which tracks are etched is called the critical angle $\theta_c$, which is equal to $\arcsin \left( \frac{V_G}{V_T} \right)$ (Fleischer and Price 1964; Fleischer et al. 1975). An etching efficiency $\eta$ is defined by the fraction of tracks intersecting a surface that are etched on the surface, and is given by $\cos^2 \theta_c$ for the case of internal “thick” sources, where particle tracks originate throughout the volume of the detector itself. Thus:

$$\rho_E = \eta \rho_L \cos^2 \theta_c = \rho_L \left( 1 - \frac{V_G^2}{V_T^2} \right)$$

where $\rho_E$ and $\rho_L$ are the areal density of etched and latent tracks respectively. $\theta_c$ can be experimentally determined by the etch-test of particle tracks that are implanted by the bombardment of collimated heavy ions at a certain angle to the sample’s surface. Such experiments yielded $\theta_c$ values of ~25-35° for natural glasses and < 10° for crystals (Khan and Durrani 1972).

When tracks originating from the internal source are etched, new tracks that began and ended beneath the original surface are revealed by progressive removal of the surface itself (Fig. 3). As a result, the total number of tracks will increase monotonically as etching proceeds because the tracks etched at an early stage continue to be visible even after long etch times, although they grow and become less distinct. This effect is called the “prolonged-etching factor” (Kahn and Durrani 1972) and is quantified by:

$$\Delta \rho = N F \rho (1 - \sin \theta_c )$$

where $\Delta \rho$ is the areal density of new tracks added, NF is a number of tracks per unit
volume and \( h \) is the thickness of the layer removed. Because \( \rho_L = gNFRL \), where \( g \) is a geometry factor of the detector (0.5 and 1 for external and internal surfaces respectively) and \( RL \) is an etchable range (i.e., length) of the latent track, thus:

\[
\Delta \rho = \rho_L \frac{\hbar(1 - \sin \vartheta_C)}{gRL} = \rho_E \frac{h}{gRL(1 + \sin \vartheta_C)}
\]

Since \( h = V_G t \), where \( t \) is a time duration of etching

\[
\Delta \rho = \rho_L \frac{V_G \left(1 - \frac{V_G}{V_T}\right)}{gRL}
\]

The total observed density of etched tracks \( \rho_0 \) is thus given by:

\[
\rho_0 = \rho_E + \Delta \rho = \rho_E \left(1 - \frac{V_G^2}{V_G^2 + V_G t} \frac{1 - \frac{V_G}{V_T}}{gRL}\right)
\]
3.5.3 Etching Criteria and Their Influences on the Observed Track Density and Length:

For reliable track density determination, the observed track density \( \rho_o \) needs to be as close to the latent track density \( \rho_L \) as possible. It is therefore desirable to analyze material that has lower \( V_G \), so that \( \rho_E \) and \( \Delta \rho \) come close to \( \rho_L \) and 0, respectively. Because \( V_G \) is anisotropic in crystals, it is necessary to employ a crystal surface having the lowest \( V_G \). This is particularly important with highly anisotropic minerals, such as zircon and sphene (Fig. 5). A conventional way to identify surfaces of low \( V_G \) is to check whether the etched surface has clear and sharp polishing scratches that indicate relatively low \( V_{Gt} \) (Naeser et al. 1980; Gleadow 1981).

Figure 3.10: (A) Shape of etched tracks in minerals having highly anisotropic VG (i.e., a big contrast between VG1 and VG2). (B) Top view of the etched mineral surface, on which track revelation is also highly anisotropic. After Gleadow (1981).

Secondly, etch time \( t \) needs to be as short as possible to minimize \( \Delta \rho \). Thus, in theory, etching should be stopped when all tracks that intersect the original surface become clearly visible under the optical microscope. Because \( V_G \) changes between individual samples, particularly zircon and sphene, the optimum etch time must be determined by the step-etch and observation procedure (Gleadow 1981; Hasebe et al. 1994). Care should be taken when etching minerals having highly anisotropic \( V_G \), on which track revelation is also anisotropic. That is, tracks lying sub-parallel to a certain crystallographic orientation (e.g., c-axis in case of zircon) are more slowly enlarged than others because they have lower rates of widening of the track channel due to lower VG perpendicular to the track orientation (Gleadow 1981). For such
samples, track etching should be continued until the tracks that are etched most slowly and weakly become visible under the microscope, so that the number of etched tracks is approximately equal for all crystallographic orientations (Gleadow 1981; Sumii et al. 1987). Otherwise, $\rho_0$ will be grossly underestimated relative to $\rho_L$. Further complexity and difficulty is due to the fact that the $V_G$ anisotropy is not constant between samples but shows a systematic decrease as a result of accumulation of radiation damages. Hence, special care is needed to etch young zircons and sphenes because they have higher anisotropy due to the low level of radiation damage and because for such samples, $\rho_L$ is in general so low that it is often difficult to judge the optimum etching condition using a small number of tracks therein (Gleadow 1980; Watanabe 1988). The observed range (i.e., length) of etched track, $R_E$, is also different from $R_L$, the etchable range of the latent track, due primarily to the outer growth of the track channel as the etching proceeds (Fig. 6). As the overestimation of the range, $\Delta R$, is approximately given by $\Delta R = 2V_Gt$ (Laslett et al. 1984), thus:

$$R_E \equiv R_L + \Delta R = R_L + 2V_Gt$$

If $V_G$ is isotropic and $V_T >> V_G$, the width of etched track, $W_E$, is approximately equal to $2V_Gt$ and is accordingly a good measure of $\Delta R$. In the case of crystals, however, $V_G$ is generally anisotropic and $W_E$ merely offers a rough estimate of $\Delta R$. For an ordinary etching condition, $\Delta R \approx W_E = \sim 1 \text{ } \mu \text{m}$ and therefore is not negligible for confined track length analysis that will be mentioned later. Since $\Delta R$ is a function of $t$ and increases as the etching proceeds, standardization of etching condition is required for reliable track length measurement as well as for inter-laboratory comparison of length data.
Figure 3.14: (A) Three stages of track revelation and associated errors ($\Delta$) of the observed track length in apatite. The dot line represents an unetched (latent) track. (B) Observed track length vs. total etching time. Note the steady increase in the observed length after being fully etched, due primarily to the outer growth of the track channel as the etching proceeds. After Laslett et al. (1984).
3.8 Track Length Measurements

Implicit in Equation (1) above is an assumption that the track lengths of spontaneous and induced tracks are the same. This is because the 3-D distribution of fission events within the apatite crystal is related to the observed track density on a 2-D surface via the average track length. In practice the lengths of spontaneous and induced tracks are never exactly the same for apatite (Gleadow et al. 1986) due to some shortening of the spontaneous tracks over their lifetime. As a result, some knowledge of the distribution of track lengths is essential in order to interpret properly the apparent fission track ages obtained. From each fission event, the two fission fragments travel in exactly opposite directions to produce a single linear damage trail with an overall length equal to the combined range of both particles. The etchable length of each track is actually somewhat shorter than the combined range as there is a small ‘range deficit’ of up to several µm at each end of the track where the damage intensity is not sufficient to produce a continuous etchable track (e.g., Fleischer et al. 1975, Iwano et al. 1993). On an internal surface of an apatite crystal tracks at all lengths up to the maximum etchable length are observed, with the shortest tracks produced from a fission event almost one fission fragment range above the surface (e.g., Gleadow and Lovering 1977). Clearly such surface-intersecting tracks are randomly truncated at some arbitrary distance along their lengths, making their use in estimating the full etchable length difficult. Several track length parameters have been utilized in fission track dating studies since the first use of projected track lengths in apatite by Wagner and Storzer (1972). They were the first to recognize that spontaneous tracks had different length distributions relative to fresh induced tracks in apatite. Dakowski (1978) established a clear understanding of the geometric properties of the various length parameters which was further extended by the work of Laslett et al. (1982). Following the work of Bhandari et al (1974) it has been shown that the greatest information about the true distribution of fission track lengths can be obtained from the measurement of horizontal “confined” tracks (Laslett et al. 1982, Gleadow et al. 1986). Confined tracks do not intersect the polished surface, but are etched wholly within the body of the mineral where the etchant has gained access below the surface along other tracks or fractures. These have been identified as Track-in-Track (TINT) or Track-in-Cleavage (TINCLE) events by Lal et al. (1969). Several examples of such
confined fission tracks, are shown in Figure 8 and procedures for measuring their lengths are described in detail by Gleadow et al. (1986).

Figure 3.11: Etched spontaneous fission tracks on a polished internal surface cut in an apatite crystal, observed at high magnification using a dry microscope objective. Most of the visible tracks are surface intersecting spontaneous tracks which are used for age determination. Arrows point to four individual confined tracks which do not intersect the surface but are fully contained within the body of the apatite crystal and etched from fractures which allow passage of etchant from the surface to the tracks. Such confined tracks are used for length measurement and provide the closest approximation to the true distribution of etchable lengths of latent, unetched fission tracks. The center pair of confined tracks also illustrate the effect of anisotropic etching with the track across the grain being much wider than the narrow track which lies closer to the elongated c-axis of the crystal. After Gleadow et al. 1986).
The application of fission track length studies to the interpretation of fission track ages depends on three properties of spontaneous fission tracks.

1. All tracks in apatite have a very similar initial length (Gleadow et al. 1986), which is controlled by the energetics of the fission decay and the nature of the track recording material (e.g., apatite).

2. Tracks become progressively shorter during exposure to elevated temperatures so that the final length is controlled principally by the maximum temperature that each track has experienced.

3. New tracks are continually added to the sample through time so that each one has experienced a different fraction of the total thermal history.

These factors combine to give a final distribution of track lengths which contains a complete record of the temperatures experienced, below about 120°C. Different length distributions result from different styles of thermal history, as illustrated in Figure 9.
3.6 Problems in Track Measurement

Measurement of the individual diffusion parameters that affect fission track annealing is not routinely undertaken. Rather, we are forced to use a proxy, the tubular hole that remains in the crystal after chemical etching of the fission damage trail. As pointed out by Crowley et al. (1991), etching involves not only removal of the damage trail itself, but also an unknown amount of the host crystal. The diameter of the etched track (~1 µm) is orders of magnitude larger than the unetched latent track (~10 nm) with a consequent loss of chemical and structural detail. Early studies of track annealing were generally based on track densities, since a clear Boltzmann-law relationship between the track density, time and temperature (Eqn. 5) had been recognized (Fleischer and Price 1964, Naeser and Faul 1969, Haack 1972). However, track density measurements (e.g., Bertel and Märk 1983) were insensitive to the subtle variations in track length that occur, particularly in samples with complex thermal histories, so that valuable information that may assist with interpretation was lost. And, although a direct relationship between length and density distributions was observed, there was considerable interest in the form of that relationship (Green 1988). Difficulties of inter-laboratory comparison of track density data also may have inhibited developments in fission track modeling (Green et al. 1988). As the method evolved, however, it became clear that confined track lengths provided greater precision in constraining annealing processes and were more amenable to modeling than track densities (Gleadow et al. 1983, 1986; Crowley 1985, Green et al. 1986). As a consequence, there has been widespread acceptance of the utility of combined track length and density measurements for fission track studies. Nevertheless, the track density method continues to find application in some specialized research fields (Carpéna 1998).

The nature of size distributions in etched track lengths is further complicated by the difficulty in determining the dimensions of unetched, or latent tracks. While there were some assumptions about the shape of a latent fission track (see Carlson 1990), their geometry has proven to be notoriously difficult to determine (Kobetich and Katz 1968). In a classic case of the act of observation modifying the observed property—radiolytic annealing was observed in apatite when samples were exposed to the electron beams applied in TEM (Silk and Barnes 1959, Paul and Fitzgerald...
1992) and microprobe analysis (Stormer et al. 1993). There has also been debate regarding the relationship of defect distribution to latent track geometry (Dartyge et al. 1981, Albrecht et al. 1982, 1985; Villa et al. 2000). Even the absolute, initial length of the latent tracks in apatite was (and remains) problematic, partly due to the very process of annealing. Fission fragment ranges have been calculated using range-energy codes (see Henk and Benton 1967, Green 1980, Crowley 1985), which produce an approximately Gaussian distribution. Ranges generated using the Ziegler et al. (1985) “SRIM” package, result in a negatively skewed distribution of fragment ranges. In an unusual annealing experiment, Donelick et al. (1990) addressed the widespread observation of earlier researchers (e.g., Wagner and Storzer 1970, Bertel et al. 1977, Green 1980, Gleadow et al. 1986) that natural (spontaneous) fission tracks are always shorter than laboratory-induced tracks. Donelick et al. (1990) conducted a series of rapid irradiation and etching experiments and showed that an initial phase of track fading occurs on a remarkably short timescale, even at room temperature. This rapid shortening impacts on the common perception that tracks are completely stable at low temperatures (Naeser 1979a). A shortening of ~0.5 µm took place at 23°C over three weeks following irradiation, but beyond this time, additional track length shortening was undetectable. Issues of precisely what is a fission track and what are its fundamental, measurable properties are not the only ones facing researchers.
3.7 Fission Track Dating Methods

The basic principles and practical methods of fission track age determination have been described elsewhere (e.g., Fleischer et al. 1975, Wagner and Van den Haute 1992, Gallagher et al. 1998) and will be summarized only briefly here. Once fission tracks have been revealed by etching, the main parameter to be measured, which is representative of geological age, is the track density or the number of tracks per unit area on the etched surface. This is measured by counting the number of track intersections with the surface using a calibrated grid in the microscope eyepiece. For a given uranium concentration, the spontaneous fission track density will steadily increase through time, provided the tracks remain stable and are therefore quantitatively retained. Determination of a fission track age requires several further experimental steps to measure the uranium concentration. The uranium concentration is not measured directly, but a second set of fission tracks is created artificially in the sample by a thermal neutron irradiation. This irradiation induces fission in a tiny fraction of the $^{235}\text{U}$ atoms, which are present in a constant ratio to $^{238}\text{U}$ in natural uranium. Knowing the total neutron fluence received during irradiation, the number of induced tracks provides a measure of the uranium concentration of the grain. Because the induced tracks are derived from a different isotope of uranium than the spontaneous tracks an important consideration in fission track dating is the assumption that the isotopic ratio of the two major isotopes of uranium, $^{235}\text{U}$ and $^{238}\text{U}$, is constant in nature. With the notable exception of the unique “natural” nuclear reactors of Oklo in Gabon (Bros et al. 1998), where this isotopic ratio is disturbed, this is a very safe assumption. Numerous measurements have shown that $^{235}\text{U}$ and $^{238}\text{U}$ are always present in their natural abundances of 0.73% and 99.27%, respectively.

The fission track age, $t$, is then calculated from the ratio of spontaneous ($\rho_s$) to induced ($\rho_i$) track densities according to the standard fission track age equation (Fleischer and Price 1964, Naeser 1967):

$$t \propto \frac{\rho_s}{\rho_i}$$
where $\lambda_D$ is the total decay constant for $^{238}\text{U}$ from all decay modes (effectively just the $\alpha$- decay constant), $\lambda_f$ is the fission decay constant, $\phi$ is the total neutron fluence received, $\sigma$ is the thermal neutron cross section for $^{235}\text{U}$, and $I$ is the $^{235}\text{U}/^{238}\text{U}$ isotopic ratio for natural uranium (effectively a constant in nature). The neutron fluence is most conveniently measured by determining the induced track density, $\rho_d$, produced in a calibrated uranium-bearing glass irradiated along with the dating samples. The fluence is related to $\rho_d$, usually measured in an mica external track detector held adjacent to the standard glass, by the following:

$$\phi = B \rho_d$$

where $B$ is a constant of proportionality. Substituting (2) into Equation (1) gives:

$$t = \frac{1}{\lambda_D} \ln \left( 1 + \frac{\lambda_D \phi \sigma I \rho_s}{\lambda_f \rho_i \rho_d} \right)$$

where

$$\zeta = \frac{\sigma IB}{\lambda_f}$$

(4)

The aggregate constant $\zeta$ (zeta) is determined empirically by measurements on age standard materials following the initial suggestion of Fleischer and Hart (1972), subsequently elaborated by Hurford and Green (1982, 1983) and Green (1985). In principle it is possible to determine the component constants individually and calculate the ages absolutely (Wagner and Van den Haute 1992) although various experimental difficulties with this approach have led to the emergence of the zeta calibration. The empirical zeta approach has been recommended by the IUGS Subcommission on Geochronology (Hurford 1990) and since adopted almost universally.
3.7.1 Experimental Procedures

Measurement of a fission track age following Equation (3) requires the determination of three different track densities, $\rho_s$, $\rho_i$ and $\rho_d$. The various different experimental strategies involved have been elaborated by Naeser (1979a), Gleadow (1981), Hurford and Green (1982) and Wagner and Van den Haute (1992) and are summarized in Figure 6. Of the five main alternatives, only the Population (PM) and

![Figure 3.12 Schematic representation of different fission track dating procedures (after Hurford and Green 1982). Of these, only population and external detector methods have gained wide currency. External Detector (EDM) methods have been extensively used for apatite and the EDM has now become the standard procedure in most laboratories. Galbraith (1984) describes the statistical treatment of analytical data derived by both these methods, and interlaboratory comparisons have mostly demonstrated excellent reproducibility using both procedures (Miller et al. 1985, 1990).

The Population Method measures both $\rho_s$ and $\rho_i$ on internal surfaces within the apatite grains themselves, but on two separate aliquots, assuming that the uranium concentrations of the two aliquots are statistically equivalent. Whereas the PM was initially the preferred method for dating apatites (e.g., Naeser 1967, Wagner 1968) the EDM is now usually preferred because it provides age information on a grain-
by grain basis. The variability between single grain ages has turned out to be important (Galbraith and Green 1990, Galbraith and Laslett 1993), as expected in the case of dating detrital apatites from sedimentary rocks, where a spread of single grain ages could be anticipated, but surprisingly, also in application to igneous rocks (e.g., O’Sullivan and Parrish 1995).

The steps involved in the External Detector Method are illustrated in Figure 7. The spontaneous tracks are etched on an internal polished surface on the apatite grains and the induced tracks on a mica external detector attached to the grain surface during neutron irradiation. After irradiation the external detector is etched to reveal an induced fission track image corresponding to grains in the apatite mount. Even though this results in a second set of tracks being produced within the apatite grains themselves, these tracks will not be detected because they are not etched after the irradiation.

Figure 3.13 The sequence of steps involved in the external detector method of fission track dating. This method is now the dominant procedure used in most fission track dating laboratories for apatite because of its ease of handling, suitability for automation and its provision of single grain age information.
3.7.2 Apatite Fission-Track Age Equations

The AFT age of a single apatite grain determined using the external detector method (EDM) is given by:

\[ t_i = \frac{1}{\lambda_d} \ln \left( 1 + \lambda_d \xi \rho_d \frac{\rho_{d,i}}{\rho_{s,i}} \right) \]

where subscript \( i \) refers to grain \( i \), \( t_i = \) fission-track age of grain \( i \), \( \lambda_d = \) total decay constant of \(^{238}\text{U} \), \( \xi = \xi \)-calibration factor based on EDM of fission-track age standards, \( g = \) geometry factor for spontaneous fission-track registration (see below), \( \rho_d = \) induced fission-track density for a uranium standard corresponding to the sample position during neutron irradiation. \( P_{s,i} = \) spontaneous fission-track density for grain \( i \) which equals \( (N_{s,i}/\Omega_i) \) where \( N_{s,i} \) is the number spontaneous fission tracks counted over area \( \Omega_i \), \( \rho_{s,i} = \) induced fission-track density for grain \( i \) which equals \( (N_{i,i}/\Omega_i) \) where \( N_{i,i} \) is the number induced fission tracks counted over area \( \Omega_i \).

Similarly, the AFT age of a single apatite grain determined using laser-ablation inductively coupled plasma-mass spectrometry (LA-ICP-MS) is given by (Donelick et al. 2005):

\[ t_i = \frac{1}{\lambda_d} \ln \left( 1 + \lambda_d \xi_{\text{MS}} \rho_{\text{MS}} \frac{\rho_{d,i}}{\rho_{i}} \right) \]

where \( P_i = (^{238}\text{U}/^{43}\text{Ca}) \) for apatite grain \( i \), \( \xi_{\text{MS}} = \xi \)-calibration factor based on LA-ICP-MS of fission-track age standards adjusted for the sample position during the LAICP-MS session and \(^{238}\text{U} = \) background-corrected \(^{238}\text{U} \) cps, \(^{43}\text{Ca} = \) backgroundcorrected \(^{43}\text{Ca} \) cps. The symmetrical error of the AFT single-grain age determined using the EDM is given by:

\[ \sigma_i = \left[ \frac{1}{N_{s,i}} + \frac{1}{N_{d,i}} + \frac{1}{N_d} + \left( \frac{\sigma_{\xi}}{\xi} \right)^2 \right]^{1/2} \]
Where \( N_d \) = number of induced fission tracks counted to determine \( \rho_d \), \( \sigma_\zeta = \) error of \( \zeta \). Similarly, the symmetrical error of the AFT single-grain age determined using LA-ICP-MS is given by:

\[
\sigma_i = \left[ \frac{1}{N_{d}} + \frac{\sigma_{PL}}{P_i} \right]^2 + \left[ \frac{\sigma_{MS}}{\zeta} \right]^2 \frac{1}{2}
\]

The pooled AFT age of a population of apatite grains determined using the EDM is given by:

\[
t_{pooled} = \frac{1}{\lambda_d} \ln \left( 1 + \lambda_d \zeta \rho_d \frac{\sum N_{ij}}{\sum N_{ij}} \right)
\]

Where \( t_{pooled} = \) the pooled AFT age. Similarly, the pooled AFT age of a population of apatite grains determined using LA-ICP-MS is given by:

\[
t_{pooled} = \frac{1}{\lambda_d} \ln \left( 1 + \lambda_d \zeta \rho_d \frac{\sum N_{ij}}{\sum P_{ij} \Omega_i} \right)
\]

The symmetrical error of the pooled AFT age determined using the EDM is given by:

\[
t_{pooled} = \left[ \frac{1}{\sum N_{ij}} + \frac{1}{\sum N_{ij}} + \frac{1}{N_d} + \left( \frac{\sigma_{\zeta}}{\zeta} \right)^2 \right]^{-1/2}
\]
3.7.2 Derivation of Age Calculation Equation

In principle, a radiometric age is given by three parameters, i.e., the numbers of parent and daughter nuclides in a material, and the decay constant for the parent nuclide. In the FT method, they are respectively the number of $^{238}\text{U}$ per unit volume, $^{238}\text{N}$, the number of spontaneous fission tracks per unit volume, $N_S$, and the decay constant for spontaneous nuclear fission,. Because $^{238}\text{U}$ also decays by α-emission with a much greater decay constant ($1.55125 \times 10^{-10} \text{ y}^{-1}$), $\lambda_D$, the basic formula for a FT age, $t$, is given by:

$$N_S = \frac{\lambda_D}{\lambda_P} N \left( \exp(\lambda_D t) - 1 \right)$$

To measure $^{238}\text{N}$, the nuclear fission reaction of $^{235}\text{U}$ that is artificially induced by thermal neutron irradiation is utilized.

The number of induced fission tracks per unit volume, $N_I$, is given by:

$$N_I = ^{235}\text{N} \sigma_F \Phi$$

where $^{235}\text{N}$ is the number of $^{235}\text{U}$ per unit volume, $\sigma_F$ is the cross section for induced nuclear fission of $^{235}\text{U}$ by thermal neutrons ($580.2 \times 10^{-24} \text{ cm}^2$), and $\Phi$ is the thermal neutron fluence. From these equations,

$$t = \frac{1}{\lambda_P} \ln \left( 1 + \frac{\lambda_D}{\lambda_F} N \left( \frac{N_I}{N} \right) I \sigma_F \Phi \right)$$

Where $I$ is the isotopic abundance of $\text{U}$, $^{235}\text{N} / ^{238}\text{N}$ ($7.2527 \times 10^{-3}$). Only tracks intersecting the etched surface are observable under an optical microscope and thus:

$$t = \frac{1}{\lambda_D} \ln \left( 1 + \frac{\lambda_D}{\lambda_F} \frac{\rho_S}{\rho_I} (QGI \sigma_F \Phi) \right)$$
where \( \rho_S \) is the surface density of etched spontaneous fission tracks, \( \rho_I \) is the surface density of etched induced fission tracks, \( Q \) is the integrated factor of registration and observation efficiency of fission tracks, and \( G \) is the integrated geometry factor of etched surface. In this equation, \( \lambda_F \) is not well determined (Bigazzi 1981; Van den haute et al. 1998), whereas \( Q \) and \( \Phi \) are generally difficult to measure accurately (Van den haute et al. 1998). Although, in principle, it is possible to determine these constants and values individually and calculate \( t \) absolutely (Wagner and Van den haute 1992; Van den haute et al. 1998), the empirical zeta calibration based on the analysis of age standards (Hurford and Green 1982, 1983; see also Fleischer et al. 1975) was recommended by the IUGS subcommission on Geochronology (Hurford 1990) and has since been used universally. \( \Phi \) is conventionally measured by the induced fission-track density on a U-doped standard glass, \( \rho_d \), irradiated together with the sample, and is thus given by:

\[
\varphi = B \rho_d
\]

where \( B \) is a calibration constant empirically determined.

Thus, the zeta age calibration factor, \( \zeta \), is defined accordingly by:

\[
\zeta = B I \sigma F / \lambda_F
\]

Hence,

\[
t = \frac{1}{\lambda_D} \ln \left( 1 + \left( \frac{\lambda_D}{\lambda_F} \right) \frac{\rho_S}{\rho_I} \right) QG \sigma F \Phi
\]

\( \zeta \) is determined empirically by analyzing a set of standard materials of known ages (Hurford 1990). \( Q \) is the same between the age standards and age-unknown samples as long as they are analyzed using identical experimental procedures and criteria. In
the routine analysis, therefore, \( Q \) is ignorable and \( \zeta \), is given by inputting the measured data of standard (i.e., \( \rho_d \), \( \rho_s \) and \( \rho_i \)) and the reference age \( t \) into the equation above.

### 3.8 Laboratory Calibration of the Apatite Fission-Track System

#### 3.8.1 General

Fission-track annealing behavior in apatite has been the subject of numerous studies since the late 1960s. The discussion here is not intended to be a comprehensive review of all of this work but is intended to illustrate the current level of understanding of this important process.

#### 3.8.2 Setting up a Calibration Procedure

The following considerations are made when setting up an AFT calibration procedure:

- Choice of apatite species or range of species for study.
- Choice of fission-track parameter(s) to be calibrated and the measurement method(s) used.
- Choice of experimental conditions to be controlled and the control methods used.
- Method or assumptions relating laboratory-derived data to data from the geological environment.
3.8.3. Choice of Apatite Species for Study
The choice of apatite species for several major studies of AFT annealing is given below:

- Durango fluorapatite (Green et al. 1986; Carlson et al. 1999; Barbarand et al. (2003a).

- Near-end-member fluorapatite and Sr-rich fluorapatite (Crowley et al 1991; Carlson et al. 1999).

- A range of Cl-contents for otherwise dominantly calcian-rich apatites (Carlson et al. 1999; Barbarand et al. 2003a).

- A range of cation substitutions with significant halogen-site substitutions (Carlson et al. 1999).

The goal or goals of the experiments should dictate the apatite species to be included in an experimental study. If the goal is to show the general nature of AFT annealing, then a study of a single apatite suffices (e.g., Green et al. 1986). If the goal is to show how Cl-content affects AFT annealing kinetics, then varying Cl with minimal variation in other parameters is desirable (e.g., Barbarand et al. 2003a). If the goal is to explore wider variations based on cation and/or anion substitutions then a range of apatite species spanning both types of substitutions can begin to address this problem (e.g., Carlson et al. 1999). Most importantly, within the limitations of the data themselves (quality, accuracy, precision) and the laboratory calibrations (accuracy, precision), all data from all sources should ultimately be reconciled in any resultant model. For a model that does not address one or another aspect of the overall data, then that model is limited, and the data excluded and reasons for excluding them should be disclosed. Fission tracks in apatite are considered to be easily studied in the laboratory. This is because apatite, unlike zircon and titanite, accumulates minimal radiation damage from the α-decay of trace amounts of uranium and thorium in its structure. As such, it is common practice to study the behavior of fission tracks formed by the thermal-neutron-induced fission of $^{235}\text{U}$ as a proxy for the behavior of spontaneous fission tracks formed by the fission of $^{238}\text{U}$. This is accomplished using the following procedures:

- Select an apatite for study.
• Heat the apatite for sufficient time at sufficient temperature to completely anneal the spontaneous fission tracks present.

• Place the apatite near the core of a nuclear reactor so that thermal neutrons can react with $^{235}\text{U}$ in the apatite and induce fission of the $^{235}\text{U}$ creating fresh, induced fission tracks.

• Subject the apatite containing thermal-neutron-induced fission tracks to a range of laboratory conditions necessary to study the fission-track annealing characteristic of interest.
3.8.2 Choice of AFT Parameter and Method of Measurement.

Experimental design can greatly affect the ability of the experimenter to successfully calibrate a parameter of interest. Several parameters are typically measured and reported in AFT calibration studies:

- The overall mean length of confined fission tracks, the standard error of the mean, and moments of the distribution about the mean (assuming Gaussian distributions) for either a) relatively high temperatures in the laboratory (generally 75-400°C; Green et al. 1986; Crowley et al. 1991; Carlson et al. 1999; Barbarand et al. 2003a or b) relatively low temperatures (essentially room temperature following induced fission track creation; Donelick et al. 1990).

- The mean lengths of confined fission tracks parallel and perpendicular to the crystallographic c-axis obtained by fitting whole or partial ellipses, the standard error of the means, and the moments of the track length distribution about the whole or partial fitted ellipses (Donelick et al. 1990; Donelick 1991; Crowley et al. 1991; Carlson et al. 1999).

- The frequency distribution of fission-track length measurements at different angles to the crystallographic c-axis (e.g., Ketcham 2003).

- The fission track density (e.g., Green 1988).

It is has been common practice in these experiments to utilize a digitizing tablet interfaced with a personal computer and use as the measurement device a projected LED image from the tablet cursor onto the microscope field-of-view via a projection tube (the exception is that only a fraction of the Green et al. 1986 data were measured this way; the remaining data were measured using a calibrated eyepiece graticule with significantly poorer resolution compared to the digitizing tablet). The precisions of individual fission-track length and angle to the c-axis measurements are approximately 0.15µm 1(σ) and 2°(σ), respectively (Donelick 1991; other researchers have not reported similar estimates but these likely apply).
3.9. AFT Data and Data Collection

3.9.1 General
There are four measurable parameter types associated with the collection of apatite fission-track data:

- Spontaneous fission-track densities are used to calculate fission-track ages of either individual grains or populations of grains.
- Relative uranium concentrations are used to calculate fission-track ages of either individual grains or populations of grains.
- Horizontal, confined fission-track lengths are used to constrain the style of cooling histories of cooling-only samples (e.g., exhumed orogenic belts) or other low-temperature phenomena including peak burial temperatures of sedimentary rocks or mineralization temperatures in ore bodies.
- Apatite fission-track annealing kinetic parameters are used to provide an estimation of the annealing behavior of individual grains or populations of grains, and allow one to group together apatite grains having similar annealing-kinetic response during later quantitative thermal-history modeling of AFT data.

These data types are described below. This is preceded, however, by a rare, but extremely important discussion of analyst bias accompanied by suggestions for minimizing this bias.

3.9.2 Spontaneous Fission-Track Densities
Spontaneous fission tracks form continuously as the result of the spontaneous fission, over geological time, of \(^{238}\)U nuclei within the host apatite crystal lattice. Fission tracks are randomly oriented and approximately linear, composed of zones of damaged crystal lattice typically less than 20 µm in length and 3 to 14 µm wide (e.g., Paul and Fitzgerald 1992). Fission tracks in this state are termed latent fission tracks and they are not visible using standard optical microscopy. To calculate a fission track age it is necessary to obtain a measure of the spatial density of these fission tracks within an apatite grain. Unlike other radiometric dating systems (e.g., U-Pb, K-Ar, Rb-Sr) for which the spatial density of radiogenic products can be determined chemically, the spatial density of fission tracks cannot be measured
chemically due to their being composed of damaged apatite. However, the damaged apatite material forming the core of the fission tracks, being at a higher free-energy state relative to the surrounding undamaged crystal and thus more chemically reactive, can be chemically etched and the resultant fission-track etch pits enlarged sufficiently for viewing using an optical microscope (Fig. 1; e.g., Fleischer et al. 1975 and references therein). For apatite, this is usually accomplished by polishing an apatite crystal to an internal surface that, with the exception of edge effects, intersects fission tracks derived equally from both above (from the material polished away) and below (from the material that remains after polishing). This geometry of fission-track registration on an apatite surface is referred to as a 4 geometry (for 4 geometry g = 0.5 in Eqs. 1a,b and 3a,b) versus the 2 geometry (g =1.0) of natural apatite grain surfaces that intersect fission tracks derived only from the apatite side. The authors prefer to etch apatite fission tracks in 5.5 M HNO3 for 20 s at 21 °C and to view the tracks using un-polarized, transmitted and/or reflected light at 1562.5× or 2000× magnification (100× dry objective, 1.25× projection tube, 12.5× or 16× oculars, respectively). The result after chemical etching is a flat surface of apatite from which emanate etched semi-tracks (partial fission tracks because a portion of each has been removed by polishing). Laslett et al. (1984) offer a detailed argument regarding the relationship between the spatial density of fission tracks in a crystal, the fission-track density on a polished and etched 4 geometry surface, and the etchable length distribution of full-length, horizontal, confined fission tracks.

The number of spontaneous fission tracks (Ns,i in Eqs. 3a and 3b) counted over a selected apatite grain area divided by the area gives the spontaneous fission-track density (ρs,i in Eqs. 1a and 1b). For large apatite grains, the area over which the spontaneous fission-track density is measured should not be closer than approximately 10 µm from the grain boundary. This is because there is a transition from 2 geometry to 4 geometry from the grain boundary inward to about 10 µm in from the grain boundary, respectively. The area counted should be free of large surface imperfections such as cracks and large etch pits and possess a minimum of spurious etch features such as inclusions and crystallographic defects. If a grain is too small to count over a meaningful area located at least 10 µm from the grain
boundaries, the whole grain can be counted provided it is reasonable to assume that the polished and etched surface is approximately halfway through the original grain and that no significant surface area is removed by polishing and etching including along the grain boundaries.

### 3.9.3 Induced Fission-Track Densities.

The EDM of AFT age analysis is a form of thermal-neutron activation, a detailed description of which is provided by Tagami and O’Sullivan (2005). To obtain relative measurements of the uranium concentration in apatite grains selected for age determination, an external detector commonly composed of low-uranium, fission-track-free muscovite mica is placed in intimate contact with the grains and the grain mount-mica pair is irradiated with thermal neutrons in the vicinity of the core of a nuclear reactor. A high thermal neutron to fast neutron ratio is preferred because thermal-neutron induced fission of $^{235}\text{U}$ is highly favored over thermal-neutron-induced fission of $^{238}\text{U}$ and $^{232}\text{Th}$; the reverse is true for fast neutrons (e.g., Fleischer et al. 1975). It is extremely difficult to confidently determine the integrated thermal-neutron flux resulting from an irradiation session. To overcome this problem, it is common practice to irradiate silicate glass standards of known and/or constant uranium concentration affixed with their own external detectors at either end of a stack of apatite grain mount samples being irradiated. For each of these uranium glasses, an induced fission-track density is measured and from these a corresponding induced fission track density is calculated for each sample position in the stack (parameter $d$ in Eqns. 1a and 3a) by linear interpolation. Examples of uranium-doped silicate glasses commonly used for EDM age analysis include Corning Glasses CN-1 through CN-6 or NIST glasses 612 or 962a. Induced fission-track densities are determined for the uranium-doped glasses irradiated along with the apatite grain mounts. The number of induced fission tracks ($N_d$ in Eqns. 2a and 4a) counted over an area on an external detector in contact with a uranium-doped glass divided by the area gives the induced fission-track density corresponding to the glass ($\rho_d$ in Eqns. 1a and 3a). These induced fission-track densities need to be measured using randomly selected areas of the external detector that was in contact with the glass. A total of 4000 induced fission tracks for $\rho_d$ determinations is a reasonable number as this total is likely to exceed the number of
spontaneous fission tracks counted in most samples and hence will not significantly limit the precision of the age measurement. Induced fission-track densities are measured on the external detectors from the apatite grain mounts and the uranium-doped glasses. Prior to removal of the external detectors from the apatite grain mounts, three widely-spaced pin-holes are punched through each external detector and into its respective apatite grain mount to serve as easily found reference points. A combination of a) the coordinates of the reference points on the apatite grain mount, b) the coordinates, in the same coordinate system as the reference points, of the apatite grains analyzed, and c) the coordinates of the reference points on the external detector permits the coordinates of the locations on the external detector corresponding to the grains analyzed to be calculated. Most laboratories use only two reference points, which permit solving the in-plane translation and rotation components of the coordinate transformation problem (two variables, two unknowns). However, a third reference point is important for a) establishing if there is a reflection component to the transformation, and b) accounting for any tilting that would otherwise degrade focus when automated analyses are arranged on other pieces of equipment such as an electron microprobe or laser ablation stage. The authors prefer etching mica detectors in 49% HF for 15 min at 23 °C to reveal fission tracks induced within the apatite grains by this irradiation and registered in the detector. The number of induced fission tracks (Ni,i in Eqns. 2a, 3a, and 4a) counted on an external detector for an apatite grain divided by the area counted gives the induced fission-track density (ρi,i in Eqn. 1a). For a large apatite grain, induced fission tracks are counted over the area of the external detector that was in intimate contact with the area of the apatite grain over which the spontaneous fission-track density was determined. For a small apatite grain for which every spontaneous fission track was counted up to the grain boundaries, every induced fission track on the external detector from that grain needs to be counted.
The measurable parameters regarding fission track lengths include:

- The complete length of each etched, horizontal, confined fission track.
- The angle of each confined fission-track relative to the crystallographic c-axis.
- The type of fission-track length measured in terms of the pathway via which the chemical etchant reached each fission track measured (i.e., TINT, TINCLE, or TINDEF).

### 39.4 Total Confined Fission-Track Lengths in Apatite Grains

It is most common to measure the lengths of etched, nominally horizontal confined fission tracks in c-axis-parallel crystallographic planes. Confined fission tracks within approximately ±10° from horizontal are acceptable candidates for length measurement because the length apparent to the analyst equals the true length multiplied by cosine of the track angle of inclination to the horizontal plane (cos[10°] equals 0.985 and this deviation from one is near the level of precision for measuring a single fission-track length; Donelick 1991). When selecting confined fission tracks for length measurement, it is essential that only confined TINT fission tracks with well-etched and clearly visible ends be considered (Donelick et al. 1990; Carlson et al. 1999; Jonckheere and Wagner 2000; restricting measurements to TINT fission tracks is discussed further below). Analyst-derived measurement bias can be minimized by adopting and using consistent criteria for selection of fission tracks for length measurement and for placement of the measuring device at the ends of each track measured. It is also very important to determine whether or not any residual fluid may be trapped in the tracks, thus lowering the relief of the track ends and potentially masking them from view (Fig. 6). If trapped fluid is apparent or suspected, the grain mount should be washed thoroughly with acetone and then dried with a bead of acetone on the surface in order to withdraw by surface tension any acetone remaining in the tracks. Acetone effectively dissolves aqueous fluid that may be entrained in the etched fission tracks. It is essential that etched apatite grain mount surfaces not be touched directly as oil from a human hand can enter the fission tracks, obscure the etched track ends, and this oil is very difficult to effectively remove with solvents. In extreme cases, immersing the grain mounts in soapy water and agitating them vigorously, followed by cleaning with acetone or
ethanol can result in the successful removal of any residual fluid from within the tracks.

3.9.5 Kinetic Parameter \( D_{\text{par}} \).

As used here, an etch figure represents the geometrical figure formed by the intersection of an etch pit (e.g., a fission track or other crystallographic imperfection) and the c-axis parallel polished and etched apatite surface. Kinetic parameter \( D_{\text{par}} \) is the mean, maximum etch figure diameter parallel to the crystallographic c-axis. Fission track etch pits in apatite, etched in 5.5 M HNO₃ for 20 s at 21 °C are elongate parallel to the c-axis and tend to exhibit hexagonal shaped etch figures (Fig. 10). The maximum diameter of one of these hexagonal etch figures is the distance between the two tips. A minimum total of four individual \( D_{\text{par}} \) values are recommended from which to calculate the mean \( D_{\text{par}} \) value although this number is not always attainable. Also, other etched features such as dislocations that intersect the surface, polishing scratches, and minute fluid inclusions can often be used for \( D_{\text{par}} \) measurement. Experience is necessary to make the distinction between an etched feature with a representative \( D_{\text{par}} \) value and one which gives a value that is too large (e.g., for a large fluid inclusion) or too small (for a feature that only began etching part-way through the etching process).

A frequent misconception on the part of fission-track workers is that parameter \( D_{\text{par}} \) is a proxy for Cl wt%. This is not the case, although it is true that \( D_{\text{par}} \) is positively correlated with Cl wt% and OH wt% and negatively correlated with F wt% in apatite (Donelick 1993; Donelick 1995; Burtner et al. 1994; Fig. 11). \( D_{\text{par}} \) is a stand-alone parameter and should be considered independent of any single chemical composition variable because a number of chemical composition variables as well as variables as yet to be fully explored or envisioned control it (Carlson et al. 1999). Some properties of kinetic parameter \( D_{\text{par}} \) follow:
Figure 3.14 Grain images and Dpar and Cl wt% values for six track length grains for a sandstone sample. Most of the etch figures visible in these grains represent fission tracks from $^{252}$Cf-derived fission-fragment irradiation.

- Apatite grains with relatively low values of Dpar ($\leq 1.75$ µm for apatite grains etched for 20 s in 5.5 M HNO3 at 21°C) anneal rapidly and can generally be considered fast-annealing, typical near-end-member calcian-fluorapatites (Carlson et al. 1999). No reliable exceptions to this statement have been observed by the authors. Apatite grains of this type are quite common.

- Apatite grains with relatively high values of Dpar (>1.75 µm for apatite grains etched for 20 s in 5.5 M HNO3 at 21°C) usually, but not always, anneal more slowly than their low Dpar counterparts (Carlson et al. 1999). Apatite grains of this type are also quite common, but they are not as common as apatite grains with relatively low values of Dpar.

- Failure of kinetic parameter Dpar is most pronounced for near-end-member hydroxyl apatite grains that exhibit high Dpar values and anneal faster than typical calcian-fluorapatites. Dpar appears to work well for near-end-member fluorapatite grains with unusual cation substitutions such as Mn, probably Fe (apparently, even in small amounts), possibly rare earth elements, and possibly some combination of OH and Cl (Carlson et al. 1999).

- Measured values of Dpar need not be converted to some equivalent Cl wt%
value (or other chemical compositional parameter) for interpretation purposes. Quite simply, apatite fission-track age and length data are best related directly to Dpar, without reference to or need of Cl wt% or other chemical composition information.

- Because of the extremely fine-scale measurements involved with Dpar, it is of critical importance that great analytical care be taken when preparing samples for measurement, especially in terms of precisely controlling etchant strength, duration, and temperature. Lack of attention to any of these parameters can easily result in poor and/or inconsistent data quality. Furthermore, it is strongly encouraged that labs interested in using Dpar conduct careful crosscalibrations (for example, using Durango and Fish Canyon apatite; e.g., Sobel et al. 2004) to compensate for subtle but inevitable variations in measurement equipment and procedures.

- It is not known exactly what controls Dpar in apatite. Certainly, chemical composition plays a role, Cl wt% being of particular apparent importance, but other parameters related to the concentration and type of crystallographic imperfections in apatite (e.g., accumulated α-particle damage, crystallization age, temperature of formation, deformation history) may also be very important.

When using $^{252}$Cf-derived fissionfragment irradiation, it is important to consider how the $^{252}$Cf-derived fission-fragment tracks mask the orientation of the crystallographic c-axis and hence give rise to potentially spurious fission track orientation measurements and estimates of Dpar. When an apatite grain mount is irradiated with $^{252}$Cf-derived fission-fragments some distance from the $^{252}$Cf source (>5 cm for a 0.5 cm diameter source), the resultant $^{252}$Cf-derived fission-fragment tracks are nearly parallel to each other and their etch figures are nearly parallel to each another as well, regardless of the true orientation of the c-axis within the target apatite grain. In such cases, it is necessary to seek out spontaneous fission tracks (from the spontaneous fission of $^{238}$U) that are randomly orientated and verify that their etch figures are parallel to one another and parallel to the $^{252}$Cf-derived etch figures. Other features such as mineral and/or fluid inclusions and the presence of a prismatic face along the apatite grain boundary can aid the identification of properly orientated grains.
3.9.6 Extrapolation of Calibrations to Geological Time

To obtain a useful mathematical model of laboratory-derived data for AFT annealing, it is only necessary that the laboratory data being fitted are defensible and that sound mathematical principles are involved with the calibration of those data. The modeler should only be concerned with whether or not the calibration predicts reasonable behavior at geological timescales and not be concerned with whether the calibration follows previously used methods. The works of Vrolijk et al. (1992) and Gleadow and Duddy (1981) provide reasonable geological benchmarks and were used by Ketcham et al. (1999) for selection of their preferred model; other benchmarks may be available now or in the future. There is no single equation form that one must be limited to and there is no published data set or calibration that one must seek to replicate.
3.10 Data Analysis and Graphical Displays

An analytical scheme for single-grain (or grain-by-grain) data obtained by the EDM is briefly presented below.

3.10.1 Statistical Test of Single-Grain Data and Error Calculation of Sample Mean Age

Routinely 10-30 single-grain ages are determined for a single FT analysis. If the grains within the sample have a common age, the variation in single grain ages is governed only by the Poissonian statistics concerned with the determination of ρS, ρI and ρD. In this case, ρS and ρI are obtained from:

\[
ρ_s = \frac{\sum N_{sj}}{\sum A_j}, \quad ρ_I = \frac{\sum N_{ij}}{\sum A_j}
\]

where \( N_{sj} \) is the number of spontaneous fission tracks in area \( A_j \) of the jth crystal and \( N_{ij} \) is the number of induced fission tracks in the same area of the corresponding grain print on the muscovite detector (e.g., Green 1981). Then, the uncertainty in the mean age \( t \) is given by the

\[
\frac{σ(t)}{t} = \left( \frac{1}{N_s} + \frac{1}{N_I} + \frac{1}{N_D} \right)^{1/2}
\]

where \( N_s, N_I \) and \( N_D \) are the total numbers of counted tracks for \( ρ_s, ρ_I \) and \( ρ_D \), respectively. In reality, however, the assumption mentioned above is not necessarily valid for a number of factors (see Green 1981) and thus a statistical procedure, called \( χ^2 \)-test (Galbraith 1981; Green 1981), was developed to assess the validity. It was shown that the results from the conventional formula give the best estimate of \( (ρ_s/ρ_I) \) and \( σ(ρ_s/ρ_I) \), as long as the observed track counts are acceptable under a \( χ^2 \)-criterion:
\[ \chi^2 = \sum \left( \frac{(N_{Sj} - P_{Sj})^2}{P_{Sj}} \right) + \sum \left( \frac{(N_{Ij} - P_{Ij})^2}{P_{Ij}} \right) \]

where \( P_{Sj} = N_S/(N_S + N_I) \); \( P_{Ij} = N_I/(N_S + N_I) \). Here, the \( \chi^2 \)-value is tested at a desirable critical level, say 5%, with \((n-1)\) degree of freedom where \( n \) is the number of grains counted.

If the \( \chi^2 \)-value is unacceptable, it suggests that the data suffer from extra-Poissonian variation(s) due to variety of experimental and geological factors (Burchart 1981; Green 1981), such as:

(a) incomplete revelation of spontaneous fission tracks, particularly in the case of zircon and sphene, which have more complex etching characteristics as mentioned above, imprecise track counting, particularly in the case of samples having high track densities or those having a great proportion of non-track features,

(b) incomplete contact between crystals and muscovite detectors,

(c) inhomogeneity of uranium within the measured grain,

(d) spatial variation of the thermal neutron flux on a scale of the areal distance of mounted grains (i.e., \( \sim 0.1 \) to \( 10 \) mm),

(e) inherent variability of single-grain ages in the sample dated, such as the case of reworked volcanic ash beds, sedimentary rocks having multiple detrital age components, and partially annealed samples with a significant variation in the thermal sensitivities among grains dated (e.g., apatites with a spread of Cl contents).

In the above cases, usage of conventional formula for the sample mean age and its uncertainty is no longer valid. Where the values of \( \log (\rho_S/\rho_I) \) can be approximated by a Normal distribution, the central age and age dispersion will yield a better approximation of the population ages (Galbraith and Laslett 1993).

(f) spatial variation of the thermal neutron flux on a scale of the areal distance of mounted grains (i.e., \( \sim 0.1 \) to \( 10 \) mm),

(g) inherent variability of single-grain ages in the sample dated, such as the case of reworked volcanic ash beds, sedimentary rocks having multiple detrital age components, and partially annealed samples with a significant variation in the thermal sensitivities among grains dated (e.g., apatites with a spread of Cl contents).
In the above cases, usage of conventional formula for the sample mean age and its uncertainty is no longer valid. Where the values of log (ρS/ρI) can be approximated by a Normal distribution, the central age and age dispersion will yield a better approximation of the population ages (Galbraith and Laslett 1993).
3.10.1 Graphical Displays of Single-Grain Age Distribution

If the extra-Poissonian variation(s) are statistically detected in the single-grain age data, the structure of the spread in measured single-grain ages should be assessed in order to constrain the source of errors. For this, graphical methods have been employed using (a) the one-dimensional plot, or histogram, (b) the interval plot using, e.g., 95% confidence intervals (Seward and Rhoades 1986), (c) the age spectra, or weighted histogram (Hurford et al. 1984), and (d) the radial plot (Galbraith 1988, 1990) (Fig. 13). Of these, the most widely used today is the radial plot, in which the uncertainty in a single age estimate is isolated so that it is easier to judge the variation in ages between crystals. When multiple age populations are deduced in the radial plot of the sample data, statistical models can be applied to estimate the component ages, particularly the youngest age population (Galbraith and Green 1990; Brandon 1992; Galbraith and Laslett 1993) (Fig. 13). This technique is critical to the study of provenance ages using the detrital FT thermochronology (see Hurford and Carter 1991; Carter and Moss 1999; Garver et al. 1999; Ruiz et al. 2004) as well as to identify and extract the “essential age” in dating volcanic ash layers (e.g., Naeser et al. 1973; Gleadow 1980; Kowallis et al. 1986; Kohn et al. 1992; Andriessen et al. 1993; O’Sullivan et al. 2001), and even granitic rocks containing apatite grains with varying compositions (O’Sullivan and Parrish 1995).

Figure. 3.15: The age spectra (top) and radial plot (bottom) of twenty apatite grains from a sample artificially composed of two age groups (i.e., ~240 and ~340 Ma) (After Galbraith and Green 1990; Wagner and Van den haute 1992). The two dashed lines show the component age estimates. In the radial plot, the horizontal and vertical axes represent the standardized age estimate and reciprocal error, respectively.
3.10.3 Graphical displays of track length distribution

Approximately 50-100 confined tracks are routinely measured for each sample, even though even this number should be considered an absolute minimum. The distribution of track lengths is classically shown using a histogram with 1 µm bins since the general overall uncertainty of measured lengths is around 0.5 µm, due primarily to the outer growth of etched track channels as mentioned above. Because the track annealing is anisotropic, the angular distribution diagram is also used widely to rigorously judge the degree of partial annealing of tracks (Green et al. 1986; Donelick 1991; Ravenhurst and Donelick 1992; Yamada et al. 1995ab; Donelick et al. 1999; Barbarand et al. 2003b) (Fig. 8). In the case of apatite, mean track lengths either parallel or perpendicular to the c-axis can be used, instead of the usual meantrack length, as fitted parameters of track annealing models (Carlson et al. 1999; Donelick et al. 1999; Ketcham et al. 1999). Mean track lengths for zircon are calculated using tracks ≥60° to c-axis so that the effects of anisotropic annealing and etching are minimized (Yamada et al. 1993, 1995ab).

The key issue in interpreting the observed length distribution is that longer tracks tend to be over-represented because they have a greater probability of being etched and observed as TINTs or TINCLEs than shorter tracks (Laslett et al. 1982) (Fig. 10). This sampling bias needs to be appropriately corrected to obtain the real length distribution, which is necessary for the quantitative thermal history modeling of the observed FT data. In theory, the bias is proportional to the track length (Laslett et al. 1982) and this was empirically confirmed by the “two-component mixtures” experiment (Galbraith et al. 1990). In practice, however, other geometrical complexities will further be introduced into the track length analysis; i.e., the anisotropy in track etching and annealing for TINTs as well as the effective thickness of host fractures for TINCLEs (Galbraith et al. 1990). Furthermore, these factors can vary according to different experimental and analytical conditions/criteria used between laboratories as well as between observers (Barbarand et al. 2003; see also Yamada et al. 1995a).

Here N is the number of defects, t is time, and c is an empirical rate constant. The constants k, h and R are Boltzmann’s, Plank’s and the gas constant respectively. The
term $Q$ is derived from published experimental data, as are the terms $A$ and $n$ in the following equation (Eqn. 8b) where $\tau$ is a dummy variable of integration over time and $l_0$ is initial track length (Carlson 1990). When Equation 8a above is combined with the equations describing an approximately Gaussian radial defect distribution, and a function linking axial reduction to varying radius, the resulting equation is:

$$r = 1 - \frac{A}{l_0} \left( \frac{k}{n} \right)^m \left[ \int_0^\infty T(\tau) \exp \left( \frac{-Q}{RT(\tau)} \right) d\tau \right]^n$$

The model attracted considerable criticism and discussion (Crowley 1993a, Carlson 1993a, Green et al. 1993, Carlson 1993b) focusing on two key areas of the model. Firstly, both took issue with the validity of the physical model and its mechanisms. It was argued that the proposed structure was not based on available physical evidence (Green et al. 1993) and that the mechanism and kinetics of the defect elimination were implausible (Crowley 1993a). Secondly, both dismissed Carlson’s (1990) model predictions as having an inadequate fit with the laboratory data sets.

Nevertheless, Carlson’s essentially semi-empirical method has gained acceptance, and has been used as the basis for further developments such as the “multi-kinetic” model of Ketcham et al. (1999). This paper is one of three (Carlson et al. 1999, Donelick et al. 1999, Ketcham et al. 1999) that have addressed many of the earlier criticisms of the Carlson (1990) model. This research group has also produced a substantial annealing data-set of mixed-compositional apatites and established a model to deal with crystallographic effects, both of which have been incorporated into their full annealing model.

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